



Light Scattering From an Optically Active Sphere Into a Circular Sensor Aperture

by

J. David Pendleton, David L. Rosen, and James B. Gillespie

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Abstract

Two methods are developed to average V and I Stokes parameters over a circular sensor aperture collecting light scattered from an optically active sphere. One method uses a two-dimensional numerical integration of Bohren's theory in scalar form, which explicitly shows the connection with Mueller matrix elements. The second method uses expansions of vector spherical harmonics in Bohren's theory to integrate over apertures of any size without convergence checks since numerical integration is avoided. Equations simplifying the analytical results for 4π scattering are obtained. Sample computations of average Stokes parameters are performed with both methods, and agreement to six decimal places is obtained.

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1. Introduction

The presence of chiral molecules in biological materials often makes these materials optically active: i.e., characterized by different complex indices of refraction for right and left circularly polarized light. A material with differences in the real part of the refractive indices is referred to as circularly birefringent, and a material with differences in the imaginary part is referred to as circularly dichroic [1]. Many optically active materials exhibit both circular birefringence (i.e., optical rotation) and circular dichroism.

Early laser light scattering research in the area of optically active particles includes the experimental measurements of Holzwarth *et al* [2] in the 1970s, who used an application of Mie theory developed by Gordon [3] to simulate scattering from spherical microorganisms. In the 1990s, measurements of Mueller matrix elements for light scattering from nonspherical microorganisms have been reported by Lofftus *et al* [4] and by Bronk *et al* [5], who also performed realistic light scattering calculations. These results provide evidence that laser-based biosensors can detect and characterize biological organisms in water or the atmosphere if experimental measurements are supported by realistic simulations.

Exact theories of light scattering from optically active particles were first given by Bohren, who gave correct solutions for spheres [1,6], spherical shells [7], and cylinders [8]. The theory for layered cylinders was later given by Kluskens and Newman [9], and the theory for spheroids was given most recently by Cooray and Ciric [10]. These theories provide algorithms for scattering in a single direction. Two Stokes parameters, referred to here as V^{Stokes} and I^{Stokes} , can be calculated as functions of the scattering direction, and the degree of circular polarization is the ratio of these Stokes parameters.

In earlier work [11], two of us (Rosen and Pendleton) applied Bohren's scattering theory in the first simulation of scattering from optically active spheres into sensor apertures. The experimental configurations considered in that work [11] are characterized by the presence of a sensor with a circular aperture. The assumption is that all light scattered into the solid angle intercepted by the circular aperture is focused onto the sensor. This means that we obtain realistic simulations of the measured signals not by computing the

Stokes parameters for a single direction but by obtaining the average of each Stokes parameter over the aperture solid angle. Our earlier paper [11] very briefly outlined the theory but emphasized a discussion of computational results, omitting any discussion of the algorithm used for the computations. To provide a better basis for future computations of this type, we discuss the theory of two relevant algorithms here. It is assumed that most readers of this article will be interested in developing computer programs for simulation of experiments, so we have taken care to give results allowing efficient computation.

The earlier computations [11] were performed with a numerical integration over the aperture solid angle. We developed this method by adapting a two-dimensional Legendre-Gaussian numerical integration algorithm originally designed by Pierce [12] for integration over a circle. The advantage of this approach over less systematic techniques is that it gives very precise and efficient results, because the location of the grid points in the aperture solid angle is optimized. A succinct description of the method is given in section 3. A computer program based on this method was developed and run on a personal computer, giving the computational results reported earlier [11]. This numerical integration method is computationally simple, but the results must be checked for convergence; i.e., the computation must be repeated with an increasing number of points in the computational grid until convergence is obtained. Also, this method cannot treat aperture solid angles approaching 2π steradians.

To overcome these limitations of the numerical integration method, we developed a second method to treat any aperture solid angle without convergence checks. This method, which does not use any numerical integration, is referred to here as the analytical method. Explicit expressions for the two average Stokes parameters are given that describe the scattering of a wave from an optically active sphere into an aperture located in any specified direction and intercepting a solid angle of any size.

The analytical method is applied here to optically active spheres for the first time. The averaging of I^{Stokes} is an easy extension of an approach developed for Mie scattering into apertures by Chylek [13], Wiscombe and Chylek [14], Chu and Robinson [15], Pendleton [16,17], and Son, Farmer, and Giel [18,19]. The averaging of V^{Stokes} with this method is new and is the aspect of this report most likely to appeal to theorists.

The theory given in these last five references used the notation of Jackson [20]. We originally developed the theory given here using Jackson's notation

but rewrote it using notation that is more standard in light scattering [1] to interface with Bohren's theory for spheres. This choice of notation should allow easy application to other particle shapes for which scattering solutions are known.

Both the numerical integration and analytical methods can treat an incident wave that is linearly polarized, randomly polarized, or unpolarized. Mathematically, a randomly polarized wave is equivalent to an unpolarized wave, so these results are referred to here as unpolarized.

The objectives of the numerical integration and analytical methods are the same, but the approaches are so different that it is surprising that the two methods give results that agree to better than six decimal places. This agreement allows one to have confidence in the precision of these methods, and the availability of two methods allows one to apply the more appropriate to a particular situation.

A computer program based on the analytical method is more computationally intensive. Such a computer program is more difficult to write and requires more computer time for small apertures than would a program for the numerical integration method. The advantage of the analytical method is that it can treat any aperture size without convergence checks.

It is hoped that application of these methods will stimulate the development of realistic simulations and new experimental techniques based on light scattering.

2. Basic Definitions

2.1 Nondimensional Stokes Parameters and the Effective Degree of Circular Polarization

For computational convenience, we define nondimensional Stokes parameters V^{Stokes} and I^{Stokes} as

$$V^{Stokes} \equiv k^2 r^2 \frac{(I_R - I_L)}{I_{inc}}, \quad (1a)$$

$$I^{Stokes} \equiv k^2 r^2 \frac{(I_R + I_L)}{I_{inc}}, \quad (1b)$$

where I_R and I_L are the irradiances of the right and left circularly polarized components of the scattered light, I_{inc} is the irradiance of the incident electromagnetic wave, and $k = 2\pi/\lambda$ is the wave number in the medium surrounding the sphere where the wavelength is λ . In the far field where I_R and I_L are inversely proportional to $k^2 r^2$, these Stokes parameters are functions of scattering direction only and are not functions of r , the distance from the sphere's center to the point where the Stokes parameters are measured.

We obtain the average Stokes parameters, $\langle V^{Stokes} \rangle$ and $\langle I^{Stokes} \rangle$, by averaging the Stokes parameters over the sensor aperture solid angle $\Delta\Omega$ with the expression

$$\langle f \rangle \equiv \frac{1}{\Delta\Omega} \int_{\Delta\Omega} d\Omega f, \quad (2a)$$

and the effective degree of circular polarization for scattering into the solid angle $\Delta\Omega$ is then defined to be the ratio of these average Stokes parameters,

$$P^{effective} \equiv \frac{\langle V^{Stokes} \rangle}{\langle I^{Stokes} \rangle}. \quad (2b)$$

This method requires that two electronic signals be measured. One of these signals is proportional to the radiant power $P^V = (I_{inc}\Delta\Omega/k^2)\langle V^{Stokes} \rangle$ and

the other is proportional to $P^I = (I_{inc}\Delta\Omega/k^2)\langle I^{Stokes} \rangle$. If the electronic response function of the system is defined to be the ratio of these two electronic signals, then the electronic response function is independent of I_{inc} and turns out to be equal to $P^{effective}$, the effective degree of circular polarization.

In bulk material irradiated by a linearly polarized beam, I_R and I_L of the transmitted wave are different only if the material is circularly dichroic. The idea motivating this research was to extend this concept from bulk to a single particle scattering into a sensor aperture so that a nonzero value of $P^{effective}$ would indicate the presence of an optically active particle. In our earlier work [11], the occurrence of a nonzero value of $P^{effective}$ for an optically inactive particle was defined to be a “false” signal of optical activity. We claimed [11] that no false signals could occur for some aperture locations and identified these aperture locations. Here, we prove this claim for both the numerical and analytical methods.

2.2 The Coordinate Systems and Euler Angles

The development of theory for scattering into arbitrarily located apertures is facilitated by the introduction of an aperture coordinate system located with respect to the beam system by Euler angles.

The convention in Mie theory, followed by Bohren in his theory of optical activity, is that an (x, y, z) Cartesian coordinate system is located with the origin at the center of the sphere, and the z -axis extends in the direction of incident wave propagation. If the incident wave is linearly polarized, the x -axis extends in the direction of the incident electrical field. The spherical system associated with the (x, y, z) Cartesian system is the (r, θ, ϕ) system with basis unit vectors $(\hat{r}, \hat{\theta}, \hat{\phi})$. To perform the integration over the aperture solid angle $\Delta\Omega$, we introduce a second coordinate system (x^A, y^A, z^A) with the same origin, but with orientation determined by the center of the aperture solid angle such that the z^A -axis extends through the center of the aperture. In the angular momentum literature, the convention is to use superscript primes on the second coordinate system. Here, the primes are replaced by A (for aperture). The two systems have the same origin, and the relative orientations are defined by Euler angles α, β , and γ as commonly used in the quantum theory of angular momentum [21,22]. The connection between the beam coordinates (x, y, z) and the aperture coordinates (x^A, y^A, z^A) is given

by the matrix equation

$$\begin{pmatrix} x^A \\ y^A \\ z^A \end{pmatrix} = R(\alpha\beta\gamma) \begin{pmatrix} x \\ y \\ z \end{pmatrix}, \quad (3a)$$

where

$$R(\alpha\beta\gamma) = R_z(\gamma)R_y(\beta)R_z(\alpha), \quad (3b)$$

$$R_z(\alpha) \equiv \begin{pmatrix} \cos \alpha & \sin \alpha & 0 \\ -\sin \alpha & \cos \alpha & 0 \\ 0 & 0 & 1 \end{pmatrix}, \quad (3c)$$

$$R_y(\beta) \equiv \begin{pmatrix} \cos \beta & 0 & -\sin \beta \\ 0 & 1 & 0 \\ \sin \beta & 0 & \cos \beta \end{pmatrix}. \quad (3d)$$

The matrix $R_z(\gamma)$ is obtained by replacement of α by γ in equation (3c). The Euler angles α and β are the ϕ and θ angles describing the direction of the z^A -axis (through the center of the aperture) as measured in the (x, y, z) system. For a circular aperture, the light scattered into the aperture is independent of the third Euler angle γ , which is a rotation about the z^A -axis. A superscript A means that the variables (r, θ, ϕ) and basis vectors $(\hat{r}, \hat{\theta}, \hat{\phi})$ of the spherical beam system are to be replaced by the variables (r, θ^A, ϕ^A) and basis vectors $(\hat{r}, \hat{\theta}^A, \hat{\phi}^A)$ of the aperture system. For a circular aperture centered on the z^A -axis, setting $d\Omega = \sin \theta^A d\theta^A d\phi^A$ with $0 \leq \phi^A \leq 2\pi$ and $\theta_{\min}^A \leq \theta^A \leq \theta_{\max}^A$ gives the integration over the aperture, where θ_{\max}^A defines the outside edge of the aperture and θ_{\min}^A defines the inside edge of the aperture. The solid angle with $\theta^A \leq \theta_{\min}^A$ is covered by a concentric “beam stop” or “beam block” so that $\Delta\Omega = 2\pi(\cos \theta_{\min}^A - \cos \theta_{\max}^A)$. As the name suggests, a beam stop is usually used to stop unscattered light of the incident laser beam from irradiating the sensor when the sensor aperture extends into the beam.

3. Numerical Integration of Stokes Parameters

In the “far-field approximation,” the scattered field is approximately transverse and may be written as $\mathbf{E} = E_\theta \hat{\theta} + E_\phi \hat{\phi}$. The irradiances of the right and left circularly polarized components of the scattered light are obtained with the relations $I_R/I_{inc} = E_R \cdot E_R^*/(E_{inc})^2$ and $I_L/I_{inc} = E_L \cdot E_L^*/(E_{inc})^2$, where $E_R = (E_\theta + iE_\phi)/\sqrt{2}$, $E_L = (E_\theta - iE_\phi)/\sqrt{2}$, and E_{inc} is the magnitude of the incident electrical field. The Stokes parameters defined in equation (1) can then be expressed as

$$V^{Stokes} = k^2 r^2 \frac{\text{Im}(2E_\theta E_\phi^*)}{(E_{inc})^2}, \quad (4a)$$

$$I^{Stokes} = k^2 r^2 \frac{[|E_\theta|^2 + |E_\phi|^2]}{(E_{inc})^2}. \quad (4b)$$

3.1 Linearly Polarized Incident Wave

Using the $\exp(-i\omega t)$ convention for the assumed monochromatic time dependence, we can represent the linearly polarized incident wave described above as $\mathbf{E}_{inc} = \hat{x}E_{inc} \exp(ikz)$, and Bohren [1] obtained the solution for the electrical field scattered from an optically active sphere as a function of the vector spherical harmonics $\mathbf{M}_{\sigma mn}^{(3)}$ and $\mathbf{N}_{\sigma mn}^{(3)}$. The superscript (3) on these vector spherical harmonics indicates that the radial function is $h_n^{(1)}(kr)$, a spherical Hankel function.

Bohren expressed his solution for the scattered electrical field as

$$\mathbf{E} = E_{inc} \sum_{n=1}^{\infty} i^n \frac{(2n+1)}{n(n+1)} \left[(ia_n)\mathbf{N}_{e1n}^{(3)} + (-id_n)\mathbf{N}_{o1n}^{(3)} + c_n\mathbf{M}_{e1n}^{(3)} + (-b_n)\mathbf{M}_{o1n}^{(3)} \right]. \quad (5)$$

The coefficients a_n and b_n reduce to the Mie coefficients with the same names when there is no optical activity. For an optically active sphere, $d_n = -c_n$, and when there is no optical activity, $d_n = -c_n = 0$.

In the far-field approximation where $h_n^{(1)}(kr) \rightarrow (-i)^{n+1} \frac{\exp(ikr)}{kr}$, the components of the scattered field \mathbf{E} may be expressed in standard form as

$$\begin{pmatrix} E\theta \\ -E\phi \end{pmatrix} = E_{inc} \frac{\exp(ikr)}{-ikr} \begin{pmatrix} S_2 & S_3 \\ S_4 & S_1 \end{pmatrix} \begin{pmatrix} \cos \phi \\ \sin \phi \end{pmatrix}, \quad (6a)$$

where the 2×2 matrix is the amplitude scattering matrix with elements S_k ($k = 1$ to 4) [1,23]. For an optically active sphere, Bohren identified the scattering matrix elements as

$$S_1 = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} (a_n \pi_n + b_n \tau_n), \quad (6b)$$

$$S_2 = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} (a_n \tau_n + b_n \pi_n), \quad (6c)$$

$$S_3 = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} (c_n \pi_n - d_n \tau_n), \quad (6d)$$

$$S_4 = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} (d_n \pi_n - c_n \tau_n), \quad (6e)$$

where π_n and τ_n are functions of $\cos \theta$ that are defined in Mie theory [1,23] (see app A). Since $d_n = -c_n$ for optically active spheres, it is found that $S_4 = -S_3$. For spheres that are not optically active, S_1 and S_2 become the functions of the same name in Mie theory and $S_4 = -S_3 = 0$. Substituting equation (6a) into equation (4a) gives

$$V^{Stokes} = S_{41} + S_{42} \cos(2\phi) + S_{43} \sin(2\phi), \quad (7a)$$

where S_{41} , S_{42} , and S_{43} are Mueller matrices defined as

$$S_{41} \equiv \text{Im}(S_1 S_3^* - S_2 S_4^*), \quad (7b)$$

$$S_{42} \equiv \text{Im}(-S_1 S_3^* - S_2 S_4^*), \quad (7c)$$

$$S_{43} \equiv \text{Im}(S_1 S_2^* - S_3 S_4^*). \quad (7d)$$

The approach used earlier [11] was to see if measurements of $\langle V^{Stokes} \rangle \neq 0$ could be used to indicate the presence of an optically active particle. Then, for spheres with no optical activity, it would be required that $\langle V^{Stokes} \rangle = 0$. For

optically inactive spheres, $S_{41} = S_{42} = 0$ but $S_{43} \neq 0$ in general. However, we show below that if $\alpha = 0$ or $\pi/2$, then $\langle S_{43} \sin(2\phi) \rangle = 0$. The result is that $\langle V^{Stokes} \rangle = 0$ for optically inactive spheres when the aperture is centered in the x - z plane ($\alpha = 0$) or the y - z plane ($\alpha = \pi/2$). These are the preferred locations to center the aperture if it is considered desirable to have $\langle V^{Stokes} \rangle = 0$ for a sphere with no optical activity, i.e., no false signal.

Substituting equation (6a) into equation (4b) gives

$$I^{Stokes} = S_{11} + S_{12} \cos(2\phi) + S_{13} \sin(2\phi), \quad (8a)$$

where S_{11} , S_{12} , and S_{13} are Mueller matrix elements defined as

$$S_{11} \equiv \frac{1}{2}(|S_1|^2 + |S_2|^2 + |S_3|^2 + |S_4|^2), \quad (8b)$$

$$S_{12} \equiv \frac{1}{2}(-|S_1|^2 + |S_2|^2 - |S_3|^2 + |S_4|^2), \quad (8c)$$

$$S_{13} \equiv \text{Re}(S_1 S_4^* + S_2 S_3^*). \quad (8d)$$

It is necessary to compute $\cos(2\phi)$ and $\sin(2\phi)$ for each point on the grid. We do this by rewriting equation (3a) as

$$\begin{pmatrix} v_1 \\ v_2 \\ v_3 \end{pmatrix} \equiv \begin{pmatrix} \sin \theta \cos \phi \\ \sin \theta \sin \phi \\ \cos \theta \end{pmatrix} = [R(\alpha\beta\gamma)]^{-1} \begin{pmatrix} \sin \theta^A \cos \phi^A \\ \sin \theta^A \sin \phi^A \\ \cos \theta^A \end{pmatrix}, \quad (9)$$

and recognizing that $\cos(2\phi) = [(v_1)^2 - (v_2)^2]/[(v_1)^2 + (v_2)^2]$. The other required expression is $\sin(2\phi) = 2v_1 v_2 / [(v_1)^2 + (v_2)^2]$. This expression can be used with equation (8) to show analytically that if $\alpha = 0$ or $\pi/2$, then $\langle V^{Stokes} \rangle = \langle S_{43} \sin(2\phi) \rangle = 0$ for a sphere with no optical activity.

Equation (2a) will now be adapted for numerical integration. A differential element of area on the flat circular aperture surface is $dA^A = \rho^A d\rho^A d\phi^A = (z^A)^2 d\Omega / \cos^3 \theta^A$ so that

$$\langle f \rangle \equiv \frac{1}{\Delta\Omega(z^A)^2} \int \int dA^A F, \quad (10)$$

where $F = f(\cos \theta^A)^3$. The Legendre-Gaussian numerical integration scheme of Pierce gives the algorithm for integrating some function F over a planar

annulus (i.e., the area between two concentric circles), and the solution to these integrals may be expressed as

$$\int \int dA^A F = A^A \sum_i \sum_j w_{ij} F_{ij}, \quad (11)$$

where the area of the annulus is $A^A = \pi[\rho_{\max}^2 - \rho_{\min}^2]$. The w_{ij} are weighting numbers normalized so that $\sum_i \sum_j w_{ij} = 1$, and the F_{ij} are the values of F at the grid points specified by Pierce's algorithm. The grid points are on concentric circles in the annulus, with the i index identifying the circles and the j index identifying the points on a particular circle. Each circle has the same number of grid points, $4n_c$, where $n_c \equiv$ the number of circles, so the total number of grid points is $4(n_c)^2$. Computations necessary for determining the point locations and the weights were done by Lowan, Davids, and Levenson [24]. The result is that

$$\langle f \rangle = \frac{\pi}{\Delta\Omega} \left(\tan^2 \theta_{\max}^A - \tan^2 \theta_{\min}^A \right) \sum_i \sum_j w_{ij} \cos^3 \theta_i^A f_{ij}. \quad (12)$$

This expression was used to compute $\langle V^{Stokes} \rangle$ and $\langle I^{Stokes} \rangle$, with equations (7a) and (8a) used to compute V^{Stokes} and I^{Stokes} at the grid points. The number of grid points required for convergence becomes larger if the scattering particle's diameter is increased, because the distribution of scattered light becomes more complex for larger particles.

3.2 Unpolarized Incident Wave

To obtain the integrands for an unpolarized incident wave most easily, we average equations (7a) and (8a) with respect to ϕ over the interval $0 < \phi < 2\pi$, producing

$$\left(V^{Stokes} \right)^U = S_{41}, \quad (13a)$$

$$\left(I^{Stokes} \right)^U = S_{11}. \quad (13b)$$

These integrands can be used in equation (11) to obtain $\langle (V^{Stokes})^U \rangle$ and $\langle (I^{Stokes})^U \rangle$. Equation (7b) shows that $S_{41} = 0$ if $S_3 = S_4 = 0$. This means that there is no false signal for any aperture position when the incident wave is unpolarized.

4. Analytical Integration of Stokes Parameters

4.1 Linearly Polarized Incident Wave

The initial step in developing the analytical theory to be given here is to rewrite equations (4) in vector form as

$$V^{Stokes} = k^2 r^2 (-i) \hat{r} \cdot (\mathbf{E} \times \mathbf{E}^*) / (E_{inc})^2, \quad (14a)$$

$$I^{Stokes} = k^2 r^2 (\mathbf{E} \cdot \mathbf{E}^*) / (E_{inc})^2. \quad (14b)$$

Equation (5) can be substituted directly into equations (14) if scattering into an aperture centered on the beam axis is the only aperture location of interest. Since we are interested in computing $\langle V^{Stokes} \rangle$ and $\langle I^{Stokes} \rangle$ for any desired aperture location, it is necessary to obtain expansions for $\mathbf{M}_{\sigma 1n}$ and $\mathbf{N}_{\sigma 1n}$ as a linear combination of vector spherical harmonics in the aperture system. Such an expansion has previously been given [16] for \mathbf{X}_{nm} , the vector spherical harmonic used by Jackson [20], and this expansion is used here to obtain expansions for $\mathbf{M}_{\sigma 1n}$ and $\mathbf{N}_{\sigma 1n}$.

The theory given here is simplified by the introduction of complex vector spherical harmonics

$$\overline{\mathbf{M}}_{mn}^{(3)} \equiv \overline{\mathbf{M}}_{emn}^{(3)} + i \overline{\mathbf{M}}_{omn}^{(3)}, \quad (15a)$$

$$\overline{\mathbf{N}}_{mn}^{(3)} \equiv \overline{\mathbf{N}}_{emn}^{(3)} + i \overline{\mathbf{N}}_{omn}^{(3)}, \quad (15b)$$

where

$$\overline{\mathbf{M}}_{\sigma mn}^{(3)} \equiv A_{mn} \mathbf{M}_{\sigma mn}^{(3)}, \quad (16a)$$

$$\overline{\mathbf{N}}_{\sigma mn}^{(3)} \equiv A_{mn} \mathbf{N}_{\sigma mn}^{(3)}, \quad (16b)$$

and

$$A_{mn} \equiv \sqrt{\frac{(2n+1)(n-m)!}{2(n+m)!}} \quad (16c)$$

is the normalizing factor for Legendre functions. The vector spherical harmonics of the beam system can now be written as a linear combination of the complex vector spherical harmonics of the aperture system. A derivation of this expansion for $\overline{\mathbf{M}}_{\sigma 1n}$ is outlined in appendix B, and the expansion of $\mathbf{M}_{\sigma 1n}$ may be written as

$$\mathbf{M}_{\sigma 1n}^{(3)} = \sum_{m=-n}^n f_{\sigma mn} \overline{\mathbf{M}}_{mn}^{(3)A}, \quad (17a)$$

where a superscript A on vector spherical harmonics indicates the use of the aperture system variables and basis vectors (see sect. 2.2). Operating on both sides of equation (16a) with $((1/k) \nabla \times)$ then gives

$$\mathbf{N}_{\sigma 1n}^{(3)} = \sum_{m=-n}^n f_{\sigma mn} \overline{\mathbf{N}}_{mn}^{(3)A}, \quad (17b)$$

where the matrix elements f_{emn} and f_{omn} are defined by the matrix equation

$$\begin{pmatrix} f_{emn} \\ f_{omn} \end{pmatrix} \equiv \frac{2}{(2n+1)} (-1)^{m+1} \exp(i\gamma) \left[\overline{\pi}_{nm}(\cos \beta) \begin{pmatrix} i \sin \alpha \\ -i \cos \alpha \end{pmatrix} + \overline{\tau}_{nm}(\cos \beta) \begin{pmatrix} \cos \alpha \\ \sin \alpha \end{pmatrix} \right]. \quad (17c)$$

To avoid an unnecessary digression, we define and discuss the functions $\overline{\pi}_{nm}$ and $\overline{\tau}_{nm}$ in appendix A.

Substituting the expansions of equations (17) into equation (5) allows Bohren's result to be rewritten as

$$\mathbf{E} = E_{inc} \sum_{n=1}^{\infty} \sum_{m=-n}^n i^n \frac{2n+1}{n(n+1)} \left[\psi_{mn}^a \overline{\mathbf{N}}_{mn}^{(3)A} + \psi_{mn}^b \overline{\mathbf{M}}_{mn}^{(3)A} \right], \quad (18a)$$

where

$$\psi_{mn}^a \equiv i(a_n f_{emn} - d_n f_{omn}), \quad (18b)$$

$$\psi_{mn}^b \equiv c_n f_{emn} - b_n f_{omn}. \quad (18c)$$

The aperture is located in the far field of the sphere. In the far-field approximation, $z_n \equiv h_n^{(1)}(kr) \rightarrow (-i)^{n+1} \frac{\exp(ikr)}{kr}$ gives the result that

$$\left(\frac{\overline{\mathbf{M}}_{mn}^{(3)A}}{\overline{\mathbf{N}}_{mn}^{(3)A}} \right) = (-i)^n \frac{\exp(ikr) \exp(\text{im}\phi^A)}{kr} \left[\hat{\theta}^A \begin{pmatrix} \overline{\pi}_{nm}(\cos \theta^A) \\ \overline{\tau}_{nm}(\cos \theta^A) \end{pmatrix} + i \hat{\phi}^A \begin{pmatrix} \overline{\tau}_{nm}(\cos \theta^A) \\ \overline{\pi}_{nm}(\cos \theta^A) \end{pmatrix} \right]. \quad (19)$$

Substituting equations (18) into equations (14) and the results into equation (2a) where the integrations over $0 \leq \phi^A \leq 2\pi$ are performed gives the result that

$$\left(\frac{\langle V^{Stokes} \rangle}{\langle I^{Stokes} \rangle} \right) = \frac{2\pi}{\Delta\Omega} \sum_{n=1}^{\infty} \sum_{n'=1}^{\infty} \frac{(2n+1)(2n'+1)}{n(n+1)n'(n'+1)} \sum_{m=-\min(n,n')}^{\min(n,n')} \left[\begin{pmatrix} -\psi_{nn'm}^{cross} \\ \psi_{nn'm}^{dot} \end{pmatrix} \overline{\mathbf{I}}_{nn'm}^{dot} + \begin{pmatrix} -\psi_{nn'm}^{dot} \\ \psi_{nn'm}^{cross} \end{pmatrix} \overline{\mathbf{I}}_{nn'm}^{cross} \right], \quad (20a)$$

where $\Delta\Omega = 2\pi(\cos \theta_{\min}^A - \cos \theta_{\max}^A)$, and

$$\psi_{nn'm}^{dot} \equiv \psi_{mn}^a \psi_{mn'}^{a*} + \psi_{mn}^b \psi_{mn'}^{b*}, \quad (20b)$$

$$\psi_{nn'm}^{cross} \equiv \psi_{mn}^a \psi_{mn'}^{b*} + \psi_{mn}^b \psi_{mn'}^{a*}, \quad (20c)$$

$$\overline{\mathbf{I}}_{nn'm}^{dot} \equiv \int_{\mu_{\max}}^{\mu_{\min}} d\mu F_{nn'm}^{dot}(\mu), \quad (21a)$$

$$\overline{\mathbf{I}}_{nn'm}^{cross} \equiv \int_{\mu_{\max}}^{\mu_{\min}} d\mu F_{nn'm}^{cross}(\mu), \quad (21b)$$

where $\mu_{\min} \equiv \cos(\theta_{\min}^A)$, $\mu_{\max} \equiv \cos(\theta_{\max}^A)$, and

$$F_{nn'm}^{dot}(\mu) \equiv \overline{\pi}_{nm}(\mu) \overline{\pi}_{n'm}(\mu) + \overline{\tau}_{nm}(\mu) \overline{\tau}_{n'm}(\mu), \quad (21c)$$

$$F_{nn'm}^{cross}(\mu) \equiv \overline{\pi}_{nm}(\mu) \overline{\tau}_{n'm}(\mu) + \overline{\tau}_{nm}(\mu) \overline{\pi}_{n'm}(\mu). \quad (21d)$$

Since $\bar{\mathbf{I}}_{nn',-m}^{dot} = \bar{\mathbf{I}}_{nn'm}^{dot}$ and $\bar{\mathbf{I}}_{nn',-m}^{cross} = -\bar{\mathbf{I}}_{nn'm}^{cross}$, equation (20a) can be rewritten as

$$\begin{pmatrix} \langle V^{Stokes} \rangle \\ \langle I^{Stokes} \rangle \end{pmatrix} = \frac{\pi}{\Delta\Omega} \sum_{n=1}^{\infty} \sum_{n'=1}^{\infty} \sum_{m=0}^{\min(n,n')} \frac{N_m(2n+1)(2n'+1)}{n(n+1)n'(n'+1)} \left[\begin{pmatrix} -\Psi_{nn'm}^{(+cross)} \\ \Psi_{nn'm}^{(+dot)} \end{pmatrix} \bar{\mathbf{I}}_{nn'm}^{dot} + \begin{pmatrix} -\Psi_{nn'm}^{(-dot)} \\ \Psi_{nn'm}^{(-cross)} \end{pmatrix} \bar{\mathbf{I}}_{nn'm}^{cross} \right], \quad (22a)$$

where N_m is the Neumann factor

$$\begin{aligned} N_m &\equiv 1 \text{ if } m = 0, \\ N_m &\equiv 2 \text{ if } m \neq 0, \end{aligned} \quad (22b)$$

and

$$\Psi_{nn'm}^{(\pm)dot} \equiv \psi_{nn'm}^{dot} \pm \psi_{nn',-m}^{dot}, \quad (22c)$$

$$\Psi_{nn'm}^{(\pm)cross} \equiv \psi_{nn'm}^{cross} \pm \psi_{nn',-m}^{cross}. \quad (22d)$$

Equations (22) can be programmed to compute the most general case for scattering of a linearly polarized incident wave.

4.2 Unpolarized Incident Wave

We obtained the solutions for an unpolarized incident wave from equations (22) by averaging over angle α to produce

$$\begin{aligned} \left(\langle V^{Stokes} \rangle \right)^U &= \frac{\pi}{\Delta\Omega} \sum_{n=1}^{\infty} \sum_{n'=1}^{\infty} \sum_{m=0}^{\min(n,n')} \frac{N_m(2n+1)(2n'+1)(-1)}{n(n+1)n'(n'+1)} \\ &\quad \left[\left(\Psi_{nn'm}^{(+cross)} \right)^U \bar{\mathbf{I}}_{nn'm}^{dot} + \left(\Psi_{nn'm}^{(-dot)} \right)^U \bar{\mathbf{I}}_{nn'm}^{cross} \right], \end{aligned} \quad (23a)$$

$$\begin{aligned} \left(\langle I^{Stokes} \rangle \right)^U &= \frac{\pi}{\Delta\Omega} \sum_{n=1}^{\infty} \sum_{n'=1}^{\infty} \sum_{m=0}^{\min(n,n')} \frac{N_m(2n+1)(2n'+1)}{n(n+1)n'(n'+1)} \\ &\quad \left[\left(\Psi_{nn'm}^{(+dot)} \right)^U \bar{\mathbf{I}}_{nn'm}^{dot} + \left(\Psi_{nn'm}^{(-cross)} \right)^U \bar{\mathbf{I}}_{nn'm}^{cross} \right], \end{aligned} \quad (23b)$$

where

$$(f(\alpha\beta\gamma))^U \equiv \frac{\int_{\alpha=0}^{2\pi} d\alpha f(\alpha\beta\gamma)}{2\pi}. \quad (23c)$$

Averaging the Ψ functions with respect to angle α gives

$$\left(\Psi_{nn'm}^{(+)\text{cross}}\right)^U = \left[\frac{4}{(2n+1)(2n'+1)}\right] i\frac{1}{2} \left[C_{nn'}^{(+)\text{cross}} - C_{n'n}^{(+)\text{cross}*}\right] F_{nn'm}^{\text{dot}}(\cos\beta), \quad (24a)$$

$$\left(\Psi_{nn'm}^{(-)\text{dot}}\right)^U = \left[\frac{4}{(2n+1)(2n'+1)}\right] i\frac{1}{2} \left[C_{nn'}^{(-)\text{dot}} - C_{n'n}^{(-)\text{dot}*}\right] F_{nn'm}^{\text{cross}}(\cos\beta), \quad (24b)$$

$$\left(\Psi_{nn'm}^{(+)\text{dot}}\right)^U = \left[\frac{4}{(2n+1)(2n'+1)}\right] \frac{1}{2} \left(C_{nn'}^{(+)\text{dot}} + C_{n'n}^{(+)\text{dot}*}\right) F_{nn'm}^{\text{dot}}(\cos\beta), \quad (24c)$$

$$\left(\Psi_{nn'm}^{(-)\text{cross}}\right)^U = \left[\frac{4}{(2n+1)(2n'+1)}\right] \frac{1}{2} \left(C_{nn'}^{(-)\text{cross}} + C_{n'n}^{(-)\text{cross}*}\right) F_{nn'm}^{\text{cross}}(\cos\beta), \quad (24d)$$

where

$$C_{nn'}^{(+)\text{cross}} = 2(a_n c_{n'}^* + d_n b_{n'}^*), \quad (25a)$$

$$C_{nn'}^{(-)\text{dot}} \equiv 2(d_n a_{n'}^* + b_n c_{n'}^*), \quad (25b)$$

$$C_{nn'}^{(+)\text{dot}} \equiv (a_n a_{n'}^* + b_n b_{n'}^* + c_n c_{n'}^* + d_n d_{n'}^*), \quad (25c)$$

$$C_{nn'}^{(-)\text{cross}} \equiv 2(a_n b_{n'}^* - d_n c_{n'}^*). \quad (25d)$$

Interchanging n and n' (a standard symmetry operation) in the $C_{n'n}^*$ terms and noting that $F_{nn'm}^{\text{cross}}(\cos\beta)$, $F_{nn'm}^{\text{dot}}(\cos\beta)$, $\bar{\mathbf{I}}_{nn'm}^{\text{dot}}$, and $\bar{\mathbf{I}}_{nn'm}^{\text{dot}}$ are unchanged after this operation allows equations (23) to be rewritten as

$$\begin{aligned}
\left(\langle V^{Stokes} \rangle\right)^U &= \\
&\frac{4\pi}{\Delta\Omega} \sum_{n=1}^{\infty} \sum_{n'=1}^{\infty} \sum_{m=0}^{\min(n,n')} \\
&\times \frac{N_m \left[\text{Im} \left(C_{nn'}^{(+)\text{cross}} \right) F_{nn'm}^{\text{dot}}(\cos \beta) \bar{\mathbf{I}}_{nn'm}^{\text{dot}} + \text{Im} \left(C_{nn'}^{(-)\text{dot}} \right) F_{nn'm}^{\text{cross}}(\cos \beta) \bar{\mathbf{I}}_{nn'm}^{\text{cross}} \right]}{n(n+1)n'(n'+1)}, \quad (26a)
\end{aligned}$$

$$\begin{aligned}
\left(\langle I^{Stokes} \rangle\right)^U &= \\
&\frac{4\pi}{\Delta\Omega} \sum_{n=1}^{\infty} \sum_{n'=1}^{\infty} \sum_{m=0}^{\min(n,n')} \\
&\times \frac{N_m \left[\text{Re} \left(C_{nn'}^{(+)\text{dot}} \right) F_{nn'm}^{\text{dot}}(\cos \beta) \bar{\mathbf{I}}_{nn'm}^{\text{dot}} + \text{Re} \left(C_{nn'}^{(-)\text{cross}} \right) F_{nn'm}^{\text{cross}}(\cos \beta) \bar{\mathbf{I}}_{nn'm}^{\text{cross}} \right]}{n(n+1)n'(n'+1)}. \quad (26b)
\end{aligned}$$

5. Reduction for Scattering into a Solid Angle 4π Steradians

We have investigated experimental configurations collecting almost all the scattered light. The light is scattered into a solid angle $\Delta\Omega$ of approximately 4π sr (i.e., $\theta_{\min}^A = 0$ and $\theta_{\max}^A = \pi$ radians = 180°). The average Stokes parameters can be calculated for this case without the added complication of a beam coordinate system and Euler angles. However, the mathematical technique required for the simplification of equations (22) and (26) when $\Delta\Omega = 4\pi$ sr may be of interest to theorists who use angular momentum theory. The result must be independent of the Euler angles since all the scattered light is collected. Further, the results for linearly polarized and unpolarized incident waves must be identical.

5.1 Linearly Polarized Incident Wave

Mathematically, the simplification of equations (22) and (26) occurs because $[\mathbf{I}_{nn'm}^{dot}]_{\mu_{\min}=1}^{\mu_{\max}=-1} = n(n+1)\delta_{nn'}$ and $[\mathbf{I}_{nn'm}^{cross}]_{\mu_{\min}=1}^{\mu_{\max}=-1} = 0$ as indicated by Arfken [22] and appendix C. The result is that

$$[\langle V^{Stokes} \rangle]_{\Delta\Omega=4\pi} = \sum_{n=1}^{\infty} \frac{(2n+1)^2}{4n(n+1)} (-1) \sum_{m=0}^n N_m \Psi_{nnm}^{(+cross)}, \quad (27a)$$

$$[\langle I^{Stokes} \rangle]_{\Delta\Omega=4\pi} = \sum_{n=1}^{\infty} \frac{(2n+1)^2}{4n(n+1)} \sum_{m=0}^n N_m \Psi_{nnm}^{(+dot)}. \quad (27b)$$

With equation (D-3) derived in appendix D, it is not difficult to show that

$$(-1) \sum_{m=0}^n N_m \Psi_{nnm}^{(+cross)} = \frac{4n(n+1)}{(2n+1)} \text{Im} [a_n c_n^* + d_n b_n^*], \quad (28a)$$

$$\sum_{m=0}^n N_m \Psi_{nnm}^{(+dot)} = \frac{1}{2} \frac{4n(n+1)}{(2n+1)} [|a_n|^2 + |b_n|^2 + |c_n|^2 + |d_n|^2]. \quad (28b)$$

These expressions give

$$\left[\langle V^{Stokes} \rangle\right]_{\Delta\Omega=4\pi} = \sum_{n=1}^{\infty} (2n+1) \text{Im} [a_n c_n^* + d_n b_n^*], \quad (29a)$$

$$\left[\langle I^{Stokes} \rangle\right]_{\Delta\Omega=4\pi} = \frac{1}{2} \sum_{n=1}^{\infty} (2n+1) [|a_n|^2 + |b_n|^2 + |c_n|^2 + |d_n|^2]. \quad (29b)$$

These are the average Stokes parameters for scattering from a linearly polarized incident wave into 4π sr.

The scattering efficiency $Q_{scattering}$ is connected to $\left[\langle I^{Stokes} \rangle\right]_{\Delta\Omega=4\pi}$, as we can see by comparing their definitions and observing that

$$Q_{scattering} = \frac{4}{(ka)^2} \left[\langle I^{Stokes} \rangle\right]_{\Delta\Omega=4\pi}. \quad (30a)$$

This gives the result that

$$Q_{scattering} = \frac{2}{(ka)^2} \sum_{n=1}^{\infty} (2n+1) [|a_n|^2 + |b_n|^2 + |c_n|^2 + |d_n|^2]. \quad (30b)$$

If there is no optical activity, then $a_n = a_n^{Mie}$, $b_n = b_n^{Mie}$, and $c_n = d_n = 0$, so that the conventional expression for Mie scattering efficiency is obtained.

5.2 Unpolarized Incident Wave

For the unpolarized wave scattering into $\Delta\Omega = 4\pi$ sr,

$$\begin{aligned} \left[\bar{\mathbf{I}}_{nn'm}^{dot}\right]_{\mu_{\max}=-1}^{\mu_{\min}=1} &= n(n+1)\delta_{nn'} \quad \text{and} \\ \left[\bar{\mathbf{I}}_{nn'm}^{cross}\right]_{\mu_{\max}=-1}^{\mu_{\min}=1} &= 0 \end{aligned}$$

give

$$\left(\langle V^{Stokes} \rangle\right)_{\Delta\Omega=4\pi}^U = \sum_{n=1}^{\infty} \frac{\text{Im} (C_{nn}^{(+cross)})}{n(n+1)} \sum_{m=0}^n N_m F_{nnm}^{dot}(\cos\beta), \quad (31a)$$

$$\left(\langle I^{Stokes} \rangle\right)_{\Delta\Omega=4\pi}^U = \sum_{n=1}^{\infty} \frac{\text{Re} (C_{nn}^{(+dot)})}{n(n+1)} \sum_{m=0}^n N_m F_{nnm}^{dot}(\cos\beta). \quad (31b)$$

With the expressions given in appendix D, it is found that

$$\sum_{m=0}^n N_m F_{nnm}^{dot}(\cos \beta) = \frac{1}{2}n(n+1)(2n+1), \quad (32)$$

so that

$$\left(\langle V^{Stokes} \rangle\right)_{\Delta\Omega=4\pi}^U = \frac{1}{2} \sum_{n=1}^{\infty} (2n+1) \operatorname{Im} \left(C_{nn}^{(+cross)} \right), \quad (33a)$$

$$\left(\langle I^{Stokes} \rangle\right)_{\Delta\Omega=4\pi}^U = \frac{1}{2} \sum_{n=1}^{\infty} (2n+1) \operatorname{Re} \left(C_{nn}^{(+dot)} \right), \quad (33b)$$

showing that

$$\left(\langle V^{Stokes} \rangle\right)_{\Delta\Omega=4\pi}^U = \left(\langle V^{Stokes} \rangle\right)_{\Delta\Omega=4\pi}, \quad (34a)$$

$$\left(\langle I^{Stokes} \rangle\right)_{\Delta\Omega=4\pi}^U = \left(\langle I^{Stokes} \rangle\right)_{\Delta\Omega=4\pi}. \quad (34b)$$

6. False Signal

In sections 3.1 and 3.2, we show that the term “false signal” (of optical activity) is equivalent to a nonzero measurement of $\langle V^{Stokes} \rangle$ for scattering from an optically inactive particle for which $c_n = -d_n = 0$.

For a linearly polarized incident wave (sect. 3.1), we show that there is no false signal for aperture locations such that $\alpha = 0$ or $\pi/2$; i.e., $\langle V^{Stokes} \rangle = 0$ for scattering from a spherical particle with no optical activity when the aperture is centered in the x - z plane or in the y - z plane. We can also obtain this result by analyzing equation (22a). If $\alpha = 0$ or $\pi/2$, or if $\beta = 0$ or π , then $\Psi_{nn'm}^{(+cross)} = \Psi_{nn'm}^{(-dot)} = 0$ for optically inactive spherical particles, so that equation (22a) gives the result that $\langle V^{Stokes} \rangle = 0$.

For an unpolarized incident beam, $C_{nn'}^{(+cross)} = C_{nn'}^{(-dot)} = 0$ for optically inactive spheres, so that equation (26a) gives the result $(\langle V^{Stokes} \rangle)^U = 0$. As in section 3.2, our results show that there is no false signal for an unpolarized incident beam.

7. Numerical Calculations

In this section, we give the results of some computations for the benefit of computational researchers who wish to verify computer programs that they develop with the theory given here. Using Mie theory, Bohren and Huffman give computed values of a_n^{Mie} and b_n^{Mie} for $\text{Re}(\widehat{N}) = 1.33$, $\text{Im}(\widehat{N}) = 1.0(10^{-8})$, and the Mie size parameter $X \equiv \pi D/\lambda = 3.0$. All the computations given here are for an optically active sphere with $X \equiv 3.0$, $\text{Re}(\widehat{N}_L) = 1.33$, $\text{Im}(\widehat{N}_L) = 2.0(10^{-8})$, $\text{Re}(\widehat{N}_R) = 1.33$, and $\text{Im}(\widehat{N}_R) = 0$. For these values, the resulting computed values of a_n , b_n , and c_n are given in table 1.

7.1 Aperture Centered on the Beam Axis

In tables 2, 3, and 4, computations of $P^{effective}$, $\langle V^{Stokes} \rangle$, and $\langle I^{Stokes} \rangle$ are given for scattering into a sensor aperture centered on the beam axis ($\alpha = \beta = 0^\circ$). For this particular aperture location, the results are identical for polarized and unpolarized incident beams.

Table 2 is for an aperture with beam block for which $\theta_{\min} = 5^\circ$ and $\theta_{\max} = 60^\circ$. We did these computations with the numerical integration scheme, giving

Table 1. Computed values of a_n , b_n , and c_n .

n	$\text{Re}(a_n)$	$\text{Im}(a_n)$	$\text{Re}(b_n)$	$\text{Im}(b_n)$
1	0.516306(10^0)	-0.499734(10^0)	0.737672(10^0)	-0.439900(10^0)
2	0.341921(10^0)	-0.474353(10^0)	0.400793(10^0)	-0.490059(10^0)
3	0.484668(10^{-1})	-0.214750(10^0)	0.935528(10^{-2})	-0.962692(10^{-1})
4	0.103458(10^{-2})	-0.321483(10^{-1})	0.688105(10^{-4})	-0.829491(10^{-2})
5	0.903755(10^{-5})	-0.300622(10^{-2})	0.283089(10^{-6})	-0.532042(10^{-3})
n	$\text{Re}(c_n)$	$\text{Im}(c_n)$		
1	-0.265630(10^{-7})	-0.717984(10^{-8})		
2	-0.261447(10^{-7})	0.697491(10^{-8})		
3	-0.202011(10^{-8})	0.611957(10^{-8})		
4	-0.268083(10^{-10})	0.662132(10^{-9})		
5	-0.186204(10^{-12})	0.526254(10^{-10})		

Table 2. $P^{effective}$, $\langle V^{Stokes} \rangle$, and $\langle I^{Stokes} \rangle$ for $\alpha = \beta = 0$, $\theta_{\min} = 5^\circ$, and $\theta_{\max} = 60^\circ$; computed with numerical integration method with identical results for polarized and unpolarized incident waves.

Number of circles	Number of points	$P^{effective}$	$\langle V^{Stokes} \rangle$	$\langle I^{Stokes} \rangle$
2	16	0.534274(10 ⁻⁷)	0.540146(10 ⁻⁶)	0.101099(10 ²)
4	64	0.499270(10 ⁻⁷)	0.700091(10 ⁻⁶)	0.140223(10 ²)
6	128	0.496561(10 ⁻⁷)	0.708648(10 ⁻⁶)	0.142711(10 ²)
8	256	0.496438(10 ⁻⁷)	0.708921(10 ⁻⁶)	0.142802(10 ²)
10	400	0.496434(10 ⁻⁷)	0.708928(10 ⁻⁶)	0.142804(10 ²)
12	576	0.496434(10 ⁻⁷)	0.709932(10 ⁻⁶)	0.142805(10 ²)
14	784	0.496434(10 ⁻⁷)	0.708928(10 ⁻⁶)	0.142804(10 ²)

Table 3. $P^{effective}$, $\langle V^{Stokes} \rangle$, and $\langle I^{Stokes} \rangle$ for $\alpha = \beta = 0$, $\theta_{\min} = 0$, and θ_{\max} as indicated; computed with analytical method with identical results for polarized and unpolarized incident waves.

$\theta_{\max} (^\circ)$	$P^{effective}$	$\langle V^{Stokes} \rangle$	$\langle I^{Stokes} \rangle$
60	0.495059(10 ⁻⁷)	0.717150(10 ⁻⁶)	0.144861(10 ²)
120	0.516809(10 ⁻⁷)	0.268489(10 ⁻⁶)	0.519513(10 ¹)
180	0.517047(10 ⁻⁷)	0.203983(10 ⁻⁶)	0.394514(10 ¹)

results for an increasing number of points to demonstrate the convergence of this method. The results given in the bottom row of table 2 were found to agree exactly with those given by the analytical method.

In table 3, computed values of $P^{effective}$, $\langle V^{Stokes} \rangle$, and $\langle I^{Stokes} \rangle$ are given for $\theta_{\min} = 0^\circ$ and θ_{\max} as indicated. These computations were made with the analytical method. The values of table 3 for $\theta_{\max} = 60^\circ$ were verified with the numerical integration method, and the computations for $\theta_{\max} = 180^\circ$ were verified with the specialized analytical result for scattering into 4π sr. Exact agreement was obtained in each case.

7.2 Off-Axis Aperture Locations

An off-axis aperture location was chosen with $\alpha = 30^\circ$ and $\beta = 60^\circ$. In table 4, computations of $P^{effective}$, $\langle V^{Stokes} \rangle$, and $\langle I^{Stokes} \rangle$ are given for $\theta_{\min} = 0^\circ$ and θ_{\max} as indicated. The computations of table 4a are for a polarized incident wave, and the computations of table 4b are for an unpolarized incident wave.

Table 4. $P^{effective}$, $\langle V^{Stokes} \rangle$, and $\langle I^{Stokes} \rangle$ for an incident wave scattering into aperture with $\alpha = 30^\circ$, $\beta = 60^\circ$, $\theta_{\min} = 0^\circ$, and θ_{\max} as indicated; computed with analytical method: (a) polarized and (b) unpolarized.

(a)			
$\theta_{\max} (^\circ)$	$P^{effective}$	$\langle V^{Stokes} \rangle$	$\langle I^{Stokes} \rangle$
60	$-0.434718(10^{-2})$	$-0.298083(10^{-1})$	$0.685693(10^1)$
120	$-0.223394(10^{-2})$	$-0.114976(10^{-1})$	$0.514676(10^1)$
180	$0.517047(10^{-7})$	$0.203983(10^{-6})$	$0.394514(10^1)$
(b)			
$\theta_{\max} (^\circ)$	$P^{effective}$	$\langle V^{Stokes} \rangle$	$\langle I^{Stokes} \rangle$
60	$0.508052(10^{-7})$	$0.349986(10^{-6})$	$0.688877(10^1)$
120	$0.512351(10^{-7})$	$0.263510(10^{-6})$	$0.514315(10^1)$
180	$0.517047(10^{-7})$	$0.203983(10^{-6})$	$0.394514(10^1)$

The values in these tables were computed with a Cray and a desktop personal computer. All results given by these two computers agreed except for the values of $P^{effective}$ and $\langle V^{Stokes} \rangle$ obtained for table 4a for $\theta_{\max} = 180^\circ$. The results given by the personal computer were incorrect in this case because of a lack of precision. Since many computations are performed on personal computers, we considered it important to explain this discrepancy. The specialized result for $\langle V^{Stokes} \rangle$ obtained for scattering into 4π sr follows from the observation that

$$\begin{aligned}
-\frac{1}{4} \sum_{m=0}^n N_m \Psi_{nnm}^{(+)\text{cross}} &= \sum_{m=0}^n N_m |f_{emn}|^2 \text{Im}(a_n c_n^*) + \sum_{m=0}^n N_m |f_{omn}|^2 \text{Im}(d_n b_n^*) \\
&\quad - \sum_{m=0}^n N_m \text{Re}[f_{emn} f_{omn}^*] [\text{Im}(d_n c_n^*) + \text{Im}(a_n b_n^*)]. \quad (35)
\end{aligned}$$

The difficulty arises when $\sum_{m=0}^n N_m \text{Re}[f_{emn} f_{omn}^*]$ is not exactly zero because this factor is multiplied by $\text{Im}(a_n b_n^*)$ where a_n and b_n are much larger than $d_n = -c_n$. Although we found theoretically that $\sum_{m=0}^n N_m \text{Re}[f_{emn} f_{omn}^*] = 0$, the limited precision of personal computers is such that if each term of $\text{Re}[f_{emn} f_{omn}^*]$ is not exactly zero, then the sum differs from zero enough to give an erroneous result when multiplied by $\text{Im}(a_n b_n^*)$. The difficulty does not arise when high-precision machines like Cray computers are used or when a personal computer is used with the factor $\text{Re}[f_{emn} f_{omn}^*] = 0$ in each term.

This factor is zero if $\alpha = 0$ or $\pi/2$, and it is also zero if $\beta = 0$ or π . Using these aperture positions corresponds to elimination of the false signal as discussed in section 6.

8. Summary

The objective of the research initiated earlier [11] was to develop a method for identifying optically active spheres by analyzing light scattered from a laser beam into a circular sensor aperture. We claimed that this could be done by computing the effective degree of circular polarization for apertures located in certain positions that generate no false signal of optical activity, and these positions were identified. Here, we prove this claim, and two methods for performing the necessary computations are developed and discussed.

We used Bohren's theory for light scattering by an optically active sphere as a starting point to develop the two methods given here, which average Stokes parameters over a circular sensor aperture located in any desired direction from the scattering sphere. The effective degree of circular polarization is defined as the ratio of two averaged Stokes parameters.

One method is based on a two-dimensional numerical integration scheme that explicitly shows the connection with Mueller matrix elements; this method is referred to as the numerical integration method. The other method is based on expansions of vector spherical harmonics and is referred to as the analytical method. The advantages and disadvantages of each method are discussed. For a dedicated programmer with a fast computer, the analytical method is preferred since it can treat apertures of any size without convergence checks. Either method can treat both polarized and unpolarized incident beams. Equations are derived that allow reduction of the general analytical expressions in the special case of scattering into 4π sr. Several tables of numerical results are given to facilitate the verification of computer codes, and we discuss potential computational difficulties with personal computers because of a lack of precision. We pointed out that these difficulties do not arise when the aperture is located in a position that generates no false signal. Mathematical details related to special functions, expansions, integrals, and reductions are given in four appendices.

Appendix A. Definition and Computation of the $\bar{\pi}_{nm}$ and $\bar{\tau}_{nm}$ Functions

The $\bar{\pi}_{nm}(\mu)$ and $\bar{\tau}_{nm}(\mu)$ functions are defined here as

$$\bar{\pi}_{nm}(\mu) \equiv \frac{m\bar{P}_{nm}(\mu)}{\sqrt{1-\mu^2}}, \quad (\text{A-1a})$$

$$\bar{\tau}_{nm}(\mu) \equiv -\sqrt{1-\mu^2}(d/d\mu)\bar{P}_{nm}(\mu), \quad (\text{A-1b})$$

where $\bar{P}_{nm}(\mu)$ is the normalized associated Legendre function as defined by Belousov [25] so that

$$\bar{P}_{nm}(\mu) \equiv A_{mn}P_{nm}(\mu), \quad (\text{A-1c})$$

where $P_{nm}(\mu) = (1-\mu^2)^{m/2} \frac{d^m}{d\mu^m} P_n(\mu)$ is the unnormalized associated Legendre function as defined by Arfken [22] and Stratton [26] and used in standard light scattering references by Bohren and Huffman [1] and van de Hulst [23]. These Legendre functions differ by a factor of $(-1)^m$ from the Legendre functions used by Jackson [20], Abramowitz and Stegun [27], and Gradshteyn and Ryzhik [28]. These last two references are of particular interest for their extensive recursion relations, but care must be exercised because of the $(-1)^m$ factor difference. The $\bar{\pi}_{nm}$ and $\bar{\tau}_{nm}$ functions are defined here to agree with Fuller's definitions [29] except with an additional factor of A_{mn} . There is a computational advantage to using normalized Legendre functions because the factorials in A_{mn} do not have to be computed separately but are easily incorporated into the recursion relations. Also, for negative values of m , the resulting functions are $\bar{P}_{n,-m}(\mu) = (-1)^m \bar{P}_{nm}(\mu)$, $\bar{\pi}_{n,-m}(\mu) = (-1)^{m+1} \bar{\pi}_{nm}(\mu)$, and $\bar{\tau}_{n,-m}(\mu) = (-1)^m \bar{\tau}_{nm}(\mu)$. These relations are simpler than those using the unnormalized Legendre functions.

We can conveniently compute the $\bar{\pi}_{nm}(\mu)$ and $\bar{\tau}_{nm}(\mu)$ functions using relations obtained by rewriting equation (8.733.1) given by Gradshteyn and Ryzhik [28] as

$$\begin{pmatrix} \mu \bar{\pi}_{nm}(\mu) \\ \bar{\tau}_{nm}(\mu) \end{pmatrix} = \left(\frac{1}{2} \right) \left[\sqrt{(n-m+1)(n+m)} \bar{P}_{n,m-1}(\mu) \right. \\ \left. \{\pm\} \sqrt{(n+m+1)(n-m)} \bar{P}_{n,m+1}(\mu) \right]. \quad (\text{A-2a})$$

If $\mu = 1$, then $\bar{P}_{nm}(1) = \sqrt{(2n+1)/2} \delta_{m,0}$ gives the result

$$\begin{pmatrix} \bar{\pi}_{nm}(1) \\ \bar{\tau}_{nm}(1) \end{pmatrix} = \frac{1}{2} \sqrt{\frac{(2n+1)n(n+1)}{2}} (\delta_{m,1} \{\pm\} \delta_{m,-1}). \quad (\text{A-2b})$$

It is helpful to realize that this expression may be rewritten as

$$\begin{pmatrix} \bar{\pi}_{nm}(1) \\ \bar{\tau}_{nm}(1) \end{pmatrix} = \frac{A_{1,n}n(n+1)}{2} (\delta_{m,1} \{\pm\} \delta_{m,-1}) \\ = \frac{(2n+1)}{4A_{1,n}} (\delta_{m,1} \{\pm\} \delta_{m,-1}). \quad (\text{A-2c})$$

Appendix B. Expansion of the Vector Spherical Harmonics

The expansion of Jackson's vector spherical harmonic can be written as [16]

$$\mathbf{X}_{nM} = \sum_{m=-n}^n D_{M,m}^{n*}(\alpha\beta\gamma) \mathbf{X}_{nm}^A, \quad (\text{B-1a})$$

where $\mathbf{X}_{nm} = \mathbf{X}_{nm}(\theta, \phi)$, $\mathbf{X}_{nm}^A = \mathbf{X}_{nm}^A(\theta^A, \phi^A)$, and

$$D_{M,m}^{n*}(\alpha\beta\gamma) = \exp(\text{im}\alpha) d_{M,m}^n(\beta) \exp(\text{im}\gamma). \quad (\text{B-1b})$$

Comparing the definitions of $\overline{\mathbf{M}}_{mn}^{(3)}$ and \mathbf{X}_{nm} gives $\overline{\mathbf{M}}_{mn}^{(3)} = \sqrt{2\pi n(n+1)} (-1)^{m+1} i z_n \mathbf{X}_{nm}$ so that equation (B-1a) can be used to obtain the relation

$$\overline{\mathbf{M}}_{\pm 1,n}^{(3)} = \sum_{m=-n}^n D_{\pm 1,m}^{n*}(\alpha\beta\gamma) (-1)^{m+1} \overline{\mathbf{M}}_{mn}^{(3)A}, \quad (\text{B-2a})$$

where

$$D_{\pm 1,m}^{n*}(\alpha\beta\gamma) = \exp(\pm i\alpha) d_{\pm 1,m}^n(\beta) \exp(\text{im}\gamma), \quad (\text{B-2b})$$

$$d_{\pm 1,m}^n(\beta) = \sqrt{\frac{2}{n(n+1)(2n+1)}} [\overline{\pi}_{nm}(\cos \beta) \pm \overline{\tau}_{nm}(\cos \beta)]. \quad (\text{B-2c})$$

The equations

$$\overline{\mathbf{M}}_{emn}^{(3)} = [\overline{\mathbf{M}}_{mn}^{(3)} + (-1)^m \overline{\mathbf{M}}_{-m,n}^{(3)}] / 2, \quad (\text{B-3a})$$

$$\overline{\mathbf{M}}_{omn}^{(3)} = [\overline{\mathbf{M}}_{mn}^{(3)} - (-1)^m \overline{\mathbf{M}}_{-m,n}^{(3)}] / (2i) \quad (\text{B-3b})$$

(with $m = 1$), and equation (B-2) then give

$$\overline{\mathbf{M}}_{\sigma 1n}^{(3)} = A_{1n} \sum_{m=-n}^n f_{\sigma mn} \overline{\mathbf{M}}_{mn}^{(3)A}, \quad (\text{B-4})$$

which can be rewritten as equations (17a) and (17b) with the expansion coefficients $f_{\sigma mn}$ given by equation (17c).

Appendix C. Evaluation of $\bar{\mathbf{I}}_{nn'm}^{dot}$ and $\bar{\mathbf{I}}_{nn'm}^{cross}$

The theory of evaluating the integrals $\bar{\mathbf{I}}_{nn'm}^{dot}$ and $\bar{\mathbf{I}}_{nn'm}^{cross}$ was mainly done by Chu and Robinson [15] with a later contribution by Son, Farmer, and Giel [18]. The results are listed here in the notation of this report for the convenience of the reader, with a few comments concerning proof of the results.

By differentiating both sides of the equation, we easily show that

$$\bar{\mathbf{I}}_{nn'm}^{cross} = \left[-m\bar{P}_{nm}(\mu)\bar{P}_{nm}(\mu) \right]_{\mu_{\max}}^{\mu_{\min}}. \quad (\text{C-1})$$

For what follows, it is helpful to rewrite the differential equation for the associated Legendre function as

$$\frac{d}{d\mu} \left(\sqrt{1-\mu^2} \bar{\tau}_{nm}(\mu) \right) = n(n+1)\bar{P}_{nm}(\mu) - \frac{m\bar{\pi}_{nm}(\mu)}{\sqrt{1-\mu^2}}. \quad (\text{C-2})$$

With this result, we can easily show (by differentiating both sides) that if $n \neq n'$, then

$$\bar{\mathbf{I}}_{nn'm}^{dot} = \left[(-1) \frac{[n(n+1)\bar{P}_{nm}(\mu)\sqrt{1-\mu^2}\bar{\tau}_{n'm}(\mu) - n'(n'+1)\bar{P}_{n'm}(\mu)\sqrt{1-\mu^2}\bar{\tau}_{nm}(\mu)]}{[n(n+1) - n'(n'+1)]} \right]_{\mu_{\max}}^{\mu_{\min}}. \quad (\text{C-3})$$

If $n = n'$, then the recursion relation

$$\bar{\mathbf{I}}_{nnm}^{dot} = \left[(-1)\bar{P}_{nm}(\mu)\sqrt{1-\mu^2}\bar{\tau}_{nm}(\mu) \right]_{\mu_{\max}}^{\mu_{\min}} + n(n+1)\bar{I}_{nnm}, \quad (\text{C-4a})$$

is obtained in the same way and used with

$$\bar{I}_{nnm} \equiv \int_{\mu_{\max}}^{\mu_{\min}} d\mu \left(\bar{P}_{nm}(\mu) \right)^2 \quad (\text{C-4b})$$

an integral computed with the supplemental recursion relation

$$\bar{I}_{nnm} = \left[\frac{\bar{P}_{nm}(\mu)\sqrt{1-\mu^2}\bar{P}_{n,m-1}(\mu)}{\sqrt{(n+m)(n-m+1)}} \right]_{\mu_{\max}}^{\mu_{\min}} + \bar{I}_{nn,m-1}. \quad (\text{C-4c})$$

Equation (C-4c) can be proved with the help of a relation, $\bar{P}_{n,m-1}(\mu) = (\mu\bar{\pi}_{nm}(\mu) + \bar{\tau}_{nm}(\mu)) / \sqrt{(n+m)(n-m+1)}$, obtained from equation (A-2a).

Appendix D. Equations Giving Reduction for Scattering into 4π Steradians

Equations required to demonstrate the simplification that occurs for scattering into 4π sr can be obtained in the following way: if $\alpha = \gamma = 0$ and $\beta \succeq 0$ and if $\theta = \phi = 0$, then $\theta^A = \beta$ and $\phi^A = \pi$ while $\hat{\theta} = -\hat{\theta}^A = \hat{x}$ and $\hat{\phi} = -\hat{\phi}^A = \hat{y}$. Substituting this set of conditions into equation (B-4) and using $\bar{\pi}_{n,1}(1) = \bar{\tau}_{n,1}(1) = A_{1n}n(n+1)/2$ from equation (A-2c) gives

$$\sum_{m=-n}^n [\bar{\pi}_{nm}(\cos \beta)]^2 = \sum_{m=-n}^n [\bar{\tau}_{nm}(\cos \beta)]^2 = \frac{1}{4}n(n+1)(2n+1), \quad (\text{D-1a})$$

$$\sum_{m=-n}^n \bar{\pi}_{nm}(\cos \beta) \bar{\tau}_{nm}(\cos \beta) = 0. \quad (\text{D-1b})$$

Equation (D-1a) can be rewritten as

$$\sum_{m=0}^n N_m [\bar{\pi}_{nm}(\cos \beta)]^2 = \sum_{m=0}^n N_m [\bar{\tau}_{nm}(\cos \beta)]^2 = \frac{1}{4}n(n+1)(2n+1), \quad (\text{D-2})$$

where N_m is the Neumann factor introduced in equation (22b). With these results, it is easy to show that

$$\sum_{m=0}^n N_m |f_{emn}|^2 = \sum_{m=0}^n N_m |f_{omn}|^2 = n(n+1)/(2n+1), \quad (\text{D-3a})$$

$$\sum_{m=0}^n N_m \text{Re}(f_{emn} f_{omn}^*) = 0. \quad (\text{D-3b})$$

Since $\bar{\pi}_{n,-m}(\mu) = (-1)^{m+1} \bar{\pi}_{nm}(\mu)$ and $\bar{\tau}_{n,-m}(\mu) = (-1)^m \bar{\tau}_{nm}(\mu)$, it is easy to show that $f_{\sigma,-m,n} = (-1)^m f_{\sigma mn}^*$. With this expression and equation (D-3), it is easy to obtain equations (28).

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